

**THE JOHNS HOPKINS UNIVERSITY  
DIELECTRICS LABORATORY**

**TR-9 Third Quarterly Progress Report  
October 1, 1963 to December 31, 1963**

**Contract No. NAS8-5253**

**Control No TP3-85286 (1F)**

**CPS-02-1130-63**

THE JOHNS HOPKINS UNIVERSITY  
DIELECTRICS LABORATORY  
405 NORTH CAROLINE STREET  
BALTIMORE 31, MARYLAND

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Title: Investigation of the Behavior of Dielectric  
Materials at High Field Strengths in a High  
Vacuum Environment

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Research Contract Director

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To: National Aeronautics and Space Administration  
George C. Marshall Space Flight Center  
Huntsville, Alabama  
Attn: PR-EC

This report was prepared by The Johns Hopkins University, Dielectrics Laboratory under Contract NAS8-5253, Investigation of the Behavior of Dielectric Materials at High Field Strengths in a High Vacuum Environment, for the George C. Marshall Space Flight Center of the National Aeronautics and Space Administration. The work was administered under the technical direction of the Propulsion and Vehicle Engineering Division, Engineering Materials Branch of the George C. Marshall Space Flight Center with E. C. McKannan, Contracting Officer's Technical Representative, acting as project manager.

# Investigation of the Behavior of Dielectric Materials at High Field Strengths in a High Vacuum Environment

## I. Mission Statement

The Contractor, as an independent Contractor and not as an agent of the Government, shall, during the term of this contract, utilize its best efforts and supply the necessary personnel, facilities, and materials (except those which may otherwise be provided), and do all things necessary for, or incident to the improvement of the basic understanding of breakdown in dielectric materials, and to provide useful engineering data, not presently available, on common dielectrics, with particular emphasis directed toward determining breakdown strengths at high frequencies.

Additional effort has been authorized to continue certain phases of previous investigation of the effects of space environment on the properties of dielectric materials. These studies are associated with the effects of a vacuum, x-ray irradiation and temperature on the a-c loss properties ( $\epsilon'$  and  $\tan\delta$ ) and the d-c conductivity of insulating materials.

## II. Factual Data

### A. D-C Conductivity

In the First and Second Quarterly Progress Reports data were presented on the conductivity of polyethylene terephthalate after vacuum exposure for periods up to 150 hours at temperatures in the range of 25 to 130°C. Because these experiments were designed to show the effects of prolonged vacuum exposure at various temperatures, each specimen was held at a fixed temperature during the entire exposure period. However, comparison of the measured values of conductivity at different temperatures after equal periods of exposure suggested that the activation energy of the conduction process had different values at temperatures above and below the glass transition range (70 - 80°C). This was shown by  $\log\sigma$  vs.  $1/T$  plots. However, it was pointed out that

these plots did not indicate the true activation energy because the points were taken from experiments on different specimens, each specimen having a different exposure history.

To determine the relationship between conductivity and temperature, measurements were made on specimens that were conditioned in vacuum at elevated temperatures (120-130°C) for 150 hours and then allowed to cool at a controlled rate. To eliminate the transient current that accompanies a change in temperature, the temperature was lowered in increments and held constant at each step until the transient had decayed. At temperatures below the glass transition region, the decay period was at least two hours. Shorter periods of 15 to 30 minutes were sufficient at temperatures above the glass transition range. Only a limited number of measurements were made at temperatures below 40°C because of the long time periods required for the transients to decay.

The results of these measurements are summarized in the  $\log \sigma$  vs.  $1/T$  curves shown in Figure 1. Typical data are given for measurements in vacuum at 180 and 320 volts and in air at 90 volts. The measurements in air were made at 90 volts in order to avoid corona.

Each of the curves of Figure 1 has a higher slope at temperatures above the glass transition region, indicating that the activation energy of the conduction process is higher at the elevated temperatures. The existence of different values of activation energy suggest that there are at least two conduction mechanisms.

Measurements at different voltages were made to determine if the current-voltage relationships were the same at temperatures above and below the transition region. The data of Figure 1 show that the conductivity is slightly higher at 320 volts than at 180 volts, but the small difference in  $\log \sigma$  is essentially constant over the entire temperature range. The dependence of conductivity on applied voltage will be examined more closely in an effort to identify the conduction mechanisms.

The measurements in air at atmospheric pressure yielded results that are essentially the same as those obtained in vacuum, as shown in Figure 1. These specimens were conditioned at 130°C for 150 hours and

then allowed to cool at a controlled rate, as described above for the vacuum conditioned specimens. It is apparent that the vacuum exposure alone does not have any great effect on conductivity. This can be seen more clearly from the curves of Figure 2, which show the effect of prolonged exposure in vacuum and in air at temperatures from 25°C to 130°C. In both environments a steady decrease in conductivity was observed during the entire exposure period. It is important to note that in each case the specimen was energized throughout the entire exposure period. This long-time decay in conductivity, which had been ascribed to the effects of prolonged vacuum exposure, is not well understood. However, it must be considered when interpreting results such as those of Figure 1, where several hours are required to complete a single series of measurements over the desired temperature range. It was found that a specimen that had been slowly cooled from 130°C to 25°C could be slowly reheated in a manner that produced the same values of conductivity as those observed during the cooling cycle. Therefore, the 150 hour preconditioning at 130°C had been long enough to provide values of conductivity that remained essentially unchanged during the period required for the measurements.

Time has not permitted a detailed analysis of all of the data that has been obtained in this phase of the program. With the addition of very recent results, which require verification, sufficient data is now available to permit such an analysis to be made.

Measurements of x-ray induced conductivity in polytetrafluoroethylene TFE-7 were also made during the present reporting period. These results are discussed in the following section.

#### B. Radiation Induced A-C Losses

In a previous program, measurements of dielectric constant and dissipation factor ( $\tan\delta$ ) were made on several materials during x-ray irradiation in high-vacuum. The largest effects were those exhibited by the polytetrafluoroethylene materials TFE-6 and TFE-7 (E. I. du Pont de Nemours and Co., Inc. designations). Large changes in dielectric constant and dissipation factor were observed during 50 KVP x-ray irradiation.

The variation of dielectric constant and  $\tan\delta$  during irradiation and during subsequent recovery in vacuum and in air (or dry nitrogen) could not be explained on the basis of known radiation effects on the structure of the tetrafluoroethylene resins.

The investigation of this effect was resumed during the second quarter of the present program. Experiments which had previously been conducted at room temperature were repeated at 89°C and 158°C. Data were given in the Second Quarterly Progress Report which showed that the increases in dielectric constant and dissipation factor were greater when the specimens were irradiated at 89°C, but were greatly reduced when the irradiation was carried out at a temperature of 158°C. Therefore, it appeared that there was a temperature in the range between 25 and 158°C at which the radiation effects were more severe.

During the present period, additional measurements were made at temperatures of 113, 130 and 162°C. In these experiments the specimens were preconditioned at the test temperature for a period of about 24 hours before the irradiation was begun. The data obtained at 100 cps are summarized in Figures 3 and 4.

Previous results have shown that the x-ray induced losses remain high during the entire exposure period if the irradiation is carried out in air at atmospheric pressure. The most significant effect of the vacuum environment is to cause a steady decrease in induced losses after a peak value is reached during the early stages of exposure. Therefore, the results shown in Figures 3 and 4 suggested that the difference in behavior at the elevated temperatures might be caused by the more effective removal of residual gases.

To determine if degassing of the specimen and the walls of the vacuum chamber was responsible for the observed temperature effect, measurements were made on specimens that were baked in vacuum at 160°C for 72 hours and then allowed to cool to room temperature in vacuum before the radiation was introduced. The moderating effect of this bake-out is shown by the data of Figures 5 and 6. The maximum values of dielectric constant and  $\tan\delta$  for the baked-out specimens were considerably less than

the corresponding values for specimens that were not baked. However, the radiation effects were still considerably greater than those observed in the high temperature experiments (Figures 3 and 4).

It was felt that more complete degassing would further reduce the induced losses. Significant quantities of gas could leak into the vacuum chamber during the period required for cooling the specimen and during the irradiation period. Because the specimen was mounted in a relatively large vacuum chamber, it took approximately 12 hours to cool from 160°C to room temperature. Another experiment was conducted in which the vacuum chamber was filled with helium during the cooling period. The increased heat transfer reduced the cooling period to about one hour. The chamber was then evacuated before the radiation was introduced.

The effects of the rapid cooling can be seen in the data of Figures 5 and 6. These specimens exhibited a rapid rise in  $\tan\delta$  and a very rapid decay. The peak value occurred at a dose of about 0.15 megarads, which is in the dose range where the smaller peaks occurred in the previous experiments summarized in Figure 3.

In all previous experiments the induced losses decayed rapidly to some low value when the irradiation was discontinued. A steady value was maintained as long as the specimens remained in vacuum. However, when the cell was filled with air or dry nitrogen (oil-pumped) an immediate increase in  $\tan\delta$  would occur and the subsequent decay would continue for many months.

In the present series of experiments it was found that after 24 hours of vacuum recovery there was no detectable change in  $\tan\delta$  when the cell was filled with helium for four hours. When the cell was then evacuated and filled with dry nitrogen, the 100 cps  $\tan\delta$  increased rapidly, reaching 0.0012 in 5 minutes, 0.016 in one hour and 0.1 in 24 hours. In a subsequent experiment the specimen was allowed to remain in the helium atmosphere for 72 hours and this caused the 100 cps  $\tan\delta$  to increase to 0.044 and dielectric constant to increase from 2.135 to 2.170.

The recovery experiments demonstrate the existence of radiation induced radicals which persist for relatively long periods in vacuum.



Recovery in vacuum for as long as 20 days does not alter the effects produced by the introduction of nitrogen or air into the test chamber. It is well known that the presence of oxygen influences the effects of radiation on the properties of TFE materials and it would be expected that the reaction that occurs when the test chamber is filled with air involves the combination of oxygen and the radicals produced by the radiation. In the case of the reaction with dry nitrogen, there is some question concerning the amount of oxygen that is present as an impurity in the commercial nitrogen gas. The slow reaction that occurs when the cell is filled with helium may also be associated with an oxygen impurity. However, the striking difference in behavior in nitrogen and helium indicates that laboratory purification of the commercial nitrogen would provide a gas of sufficient purity to determine if nitrogen is involved in the reaction.

In all of the irradiation experiments with TFE materials the x-ray induced losses decrease rapidly with increasing frequency. Typical data can be found in Table I of the Second Quarterly Progress Report. This type of frequency response suggests that an induced conduction mechanism is responsible for the high losses at 100 cps. Meaningful measurements of induced d-c conductivity had not been made in the previous program because it was not possible to schedule such tests. In the present reporting period experiments were initiated to determine the effects of prolonged exposure to x-ray irradiation in vacuum on the d-c conductivity of TFE. 7.

Figure 7 shows the preliminary data obtained at 23°C using a dose rate of  $2.2 \times 10^5$  rads/hour. The induced conductivity follows the same general pattern as the induced losses at 100 cps. The recovery in vacuum is shown in Figure 8. When the cell was filled with dry nitrogen an immediate increase in conductivity was observed, just as an increase in  $\tan \delta$  occurs under similar conditions.

Further investigation will be required to determine the extent to which induced conductivity accounts for the induced a-c losses. This study will be continued during the next reporting period.

C. Calorimetric Measurement of Dielectric Losses at High Electrical Stresses in the Radio Frequency Range

(1) Introduction.

Details of a calorimetric method of measuring dielectric losses at high electrical stresses in the radio frequency range were given in the First Quarterly Progress Report. Briefly, the principle of operation consists of measuring the heat flow along a known heat conductor which is in contact with the specimen at one end and in contact with a heat sink at its other end. The specimen is placed between two such heat leads that also serve as electrodes. The opposite end of each heat lead is bonded to a constant temperature ( $0^{\circ}\text{C}$ ) heat sink. Contact to the specimens is made by a flat plate that is bonded to the end of each tubular heat lead.

A thermistor imbedded in one of the end plates is used to measure the temperature difference between the end plate and the heat sink. When an r-f voltage is applied, the temperature of the specimen will increase to some stable value. In the steady state, no additional heat is being stored in the specimen, so the measured heat flow along the heat leads is a measure of the power dissipated in the specimen.

Details of the calibration and the method of computing loss factor can be found in the First Quarterly Progress Report.

The problems that were encountered during preliminary measurements and the modifications in techniques that have been devised are presented in the following section.

(2) Limitations Imposed by the Use of Continuous Voltages at 2 Mc.

When dielectric losses are measured as a function of field strength with this type of calorimeter, the voltage is increased in steps and sufficient time must be allowed at each step for the temperature of the specimen to stabilize. It was found that periods of 10 to 30 minutes were required, depending on the magnitude of the incremental increase in applied voltage. This relatively long-time application of voltage imposes a limitation on the

maximum stress that can be used without causing the specimen to fail. With polystyrene, for example, measurements on 10 mil specimens could not be made at stresses exceeding 400 VPM.

A second limitation is encountered when measurements are to be made over a wide range of stresses. If, for example, it is desirable to make measurements from 30 to 1800 VPM, a ratio of 1:60, the power dissipated in the specimen would have a range of 1:3600. To cover such a range, major adjustments would have to be made in the conductance of the heat leads to prevent intolerable heating of the specimen. Therefore, it would be necessary to interrupt an experiment to replace heat leads whenever the temperature of the specimen reached a predetermined limit. The experiment itself would become more difficult because uniform thermal contact between the heat lead and the specimen becomes more important as the conductance of the heat lead is increased.

Another source of difficulty at high stresses is associated with the design of a suitable heat sink. The power dissipation in a typical polystyrene specimen at 2 Mc is about 0.2 watts for an applied stress of 100 VPM, but at 1800 VPM it would be 65 watts. A suitable heat sink would have to absorb this power without allowing a temperature rise of more than  $0.1^{\circ}\text{C}$  at the end of the heat lead that is bonded to the heat sink.

These difficulties, which are encountered when continuous r-f voltages are used, can be avoided by drastically reducing the time of voltage application. The benefits of using short pulses, rather than continuous voltages, are twofold: (a) the specimen can withstand a higher voltage as demonstrated by the electric strength data in the Second Quarterly Progress Report; and (b) excessive power dissipation in the specimen is avoided.

Consideration was given to a single-pulse technique, but the difficulties associated with the transient heat measurement makes this method unattractive for the purposes of the present investigation. Instead, a method was developed in which repeated r-f pulses are applied to the specimen. The repetition rate and pulse width are not critical as long as the temperature fluctuations remain negligible. The maximum stress at

which measurements can be made is lower than it would be for the single-pulse method, but it is considerably higher than for the continuous voltage method that was used in the earlier experiments.

### (3) Repeated Pulse Method.

Using the repeated-pulse method, an initial measurement of power dissipated per unit volume (Watts/cm<sup>3</sup>) is made by applying a continuous r-f field at some low stress  $E_1$ . The technique used in this first step and the method of calculating  $\tan \delta$  from the experimental data were discussed in the First Quarterly Progress Report. The stress is then increased to a value  $E_2$ , but it is now applied in repeated pulses, with a repetition rate and pulse width that result in the same average power dissipation as the continuously applied stress  $E_1$ . The average power dissipation,  $W$ , depends on the ratio  $\alpha/\beta$ , where  $\alpha$  is the pulse duration and  $\beta$  is the time from the beginning of one pulse to the beginning of the next pulse, i. e., the reciprocal of the repetition rate.

The stress is then increased to successively higher levels and at each step the ratio  $\alpha/\beta$  is adjusted so that the average power dissipation remains the same. This can be accomplished by changing  $\alpha$ ,  $\beta$  or both  $\alpha$  and  $\beta$ .

The instantaneous power dissipation,  $W'$ , is given by

$$W' = (\beta/\alpha) W$$

It is  $W'$  that is a measure of the loss factor  $\epsilon''$ , and if  $\epsilon''$  is independent of the applied stress,  $E$ , then  $W'$  should be proportional to  $E^2$ .

To determine if  $\epsilon''$  is a function of  $E$ , it is only necessary to measure the values of  $E$ ,  $\alpha$  and  $\beta$  that result in a constant value  $W$  at each step in the stress range of interest. At any two stress levels,  $E_n$ ,  $E_m$ , the following relations hold:

$$W'_m = (\beta_m/\alpha_m) (\alpha_n/\beta_n) W'_n \quad (1)$$

$$\text{and} \quad W'_m/W'_n = (E_m/E_n)^2, \text{ for constant } \epsilon''. \quad (2)$$

$$\text{Since} \quad (E_m/E_n)^2 (W'_n/W'_m) = 1,$$

then, substituting from equation (1),

$$(E_m/E_n)^2 (\beta_n/\alpha_n) (\alpha_m/\beta_m) = 1. \quad (3)$$

If  $\epsilon''$  is a function of  $E$ , then the measured values of  $E$ ,  $\alpha$  and  $\beta$  do not yield a value of unity in equation (3).

In the former method, where continuous stresses were applied, a considerable length of time was required at each stress level for the temperature of the specimen to stabilize. In the present method, this will occur only for the first step, where the stress is low. From there on, with some experience in adjusting  $\alpha$  and  $\beta$ , the temperature fluctuations can be kept small and the time required for temperature stabilization is drastically reduced.

Because the specimen temperature is adjusted to the same value throughout the experiment, the calorimeter serves only as a sensitive temperature difference indicator. The average thermodynamic conditions remain the same at each stress level, so the inherent errors in the calorimeter have no significant effect on the ratios of the measured loss factors. The largest error is made in the measurement of the actual value of  $\epsilon''$  in the first step of the experiment. This error could be reduced by constructing a calorimeter that is designed specifically for measurements at low stresses.

#### (4) Preliminary Measurements.

Preliminary measurements have been made on the following materials:

PS-C	Polystyrene, obtained from local supplier, undetermined origin.
PS-D	Polystyrene. Dow Chemical Company, Styron 666.
PE	Polyethylene. Du Pont Alathon 10
FEP	Copolymer of tetrafluoroethylene and hexafluoropropylene. Du Pont FEP-100.
C-1147	Methyl styrene with siloxane additive. Delaware Research and Development Corporation.

The detailed results of eight experiments are given in Table I. The values of  $\tan\delta$  are those that were determined in the first step of the experiment at a continuous stress. The ratio of loss factors  $\epsilon''_n/\epsilon''_m$ , at the stress levels  $E_m$  and  $E_n$  are given for each subsequent step in the

experiment. The actual power dissipation in watts is shown for each stress level. In these experiments the shortest pulse duration was about 2 milliseconds and the smallest value of  $\alpha/\beta$  was about 0.009.

The advantage of the repeated pulse method is demonstrated by the high values of stress that were used. With polystyrene, for instance, a stress of over 1000 VPM did not cause breakdown or excessive heating of the specimen. Previously, using continuous voltage application, measurements could not be made at stresses greater than 400 VPM. The present limitation is associated with the r-f generator, rather than the calorimeter. A considerable amount of time has been devoted to modifying the existing equipment to obtain properly shaped pulses of sufficiently short duration. It has been difficult to control the transients and instabilities at high voltages. The overall accuracy of the measurements is reduced if the r-f pulses deviate from a simple rectangular shape. Comparison of the ratios of  $\epsilon''$  at similar stresses for polystyrene specimens 2 and 3 of Table I, or the ratios of  $\epsilon''$  for a given specimen at the same stress levels, but at different values of power dissipation, indicate that the accuracy of the measurements must be improved. The pulse shape is critical because the instantaneous power dissipation is proportional to the square of the applied voltage.

The high values of loss that were observed at the highest stress levels can be associated with the presence of discharges that contribute to the measured power. At these high levels of stress the problems encountered in the fabrication of specimens and in the application of electrodes are similar to those for breakdown tests. The actual test area of the loss specimen must be larger than for the breakdown specimen so that the error caused by heat generated at the edges of the test area can be reduced to a negligible value. The larger area reduces the electric strength of the specimen and is also more difficult to machine. Details of the breakdown and loss measurement specimens have been given in the previous Quarterly Progress Reports.

In the case of breakdown tests, discharges that are located away from the test area do not necessarily interfere with the measurement. With the loss specimens, however, any discharges that contribute to the

measured power cause an error. The high value shown in Table I for polyethylene (specimen 7), for instance, was caused by heavy discharges from the edge of the ground electrode. It is desirable, therefore, to make the loss measurements at low voltages using thinner specimens to attain higher stresses. The thinner sections are, however more difficult to machine, so the ultimate limitation of the method will be in the ability to fabricate suitable specimens.

No conclusions can be drawn from these preliminary measurements. Further refinement in the experimental techniques seems to be justified and will be made in the next reporting period. Efforts are now being made to improve the performance of the r-f generator under repeated pulse operating conditions so that loss measurements and breakdown measurements can be made at higher stresses.

### III. Anticipated Work

During the next quarterly period the following work is planned:

- A. A continuation of the study of the effects of temperature, vacuum and irradiation on the conductivity of polymers.
- B. Further investigation of x-ray induced a-c losses in tetrafluoroethylene polymers.
- C. A continuation of the loss measurements at high field strengths and breakdown measurements in the r-f range.
- D. The investigation of loss properties at high temperatures in the r-f range.

### IV. Level of Effort

The total level of effort during this quarter has been 19.5 man-months.

The funds that were originally made available for this program have been expended and plans have been made for a continuation of the research work at approximately the same level of effort. A Revised Program Planning Chart will be submitted after the details of the contract extension have been confirmed.



**Table I. Detailed Results of Calorimeter Measurements at 2 Mc.**

Specimen#	Material	Stress Level (VPM)		$(\epsilon''_n / \epsilon''_m)$	tan $\delta$	Average Power (Watts)
		$E_m$	$E_n$			
1	PS-C (10 mil)	178	---	---	0.0004	0.4
		178	230	1.01		0.4
		178	280	0.92		0.4
		178	387	0.94		0.4
		178	429	1.09		0.4
		309	633	1.16		1.1
2	PS-D (10 mil)	175	---	---	0.0006	0.6
		175	232	1.00		0.6
		232	286	0.94		0.6
		286	393	1.03		0.6
		393	588	0.91		0.6
		226	400	0.92		1.0
		400	562	1.04		1.0
		562	734	1.18		1.0
3	PS-D (10 mil)	114	---	---	0.0004	0.15
		114	173	0.95		0.15
		173	264	1.02		0.15
		264	375	1.05		0.15
		375	545	1.07		0.15
		545	788	1.06		0.15
		340	820	1.03		0.47
		820	1030	1.40		0.47
4	C-1147 (10 mil)	146	---	---	0.0005	0.30
		146	228	0.96		0.30
5	C-1147 (30 mil)	102	---	---	0.0005	0.21
		102	202	1.05		0.21
		202	247	1.05		0.21
		247	380	0.99		0.21
6	FEP (30 mil)	88	---	---	0.0006	0.25
		88	215	1.05		---
7	PE (30 mil)	190	250	1.07	---	0.12
		250	390	1.9*		---
8	PE (20 mil)	125	---	---	0.0002	0.18
		125	170	0.95		0.18

\* caused by excessive heating due to discharges from edge of ground electrode

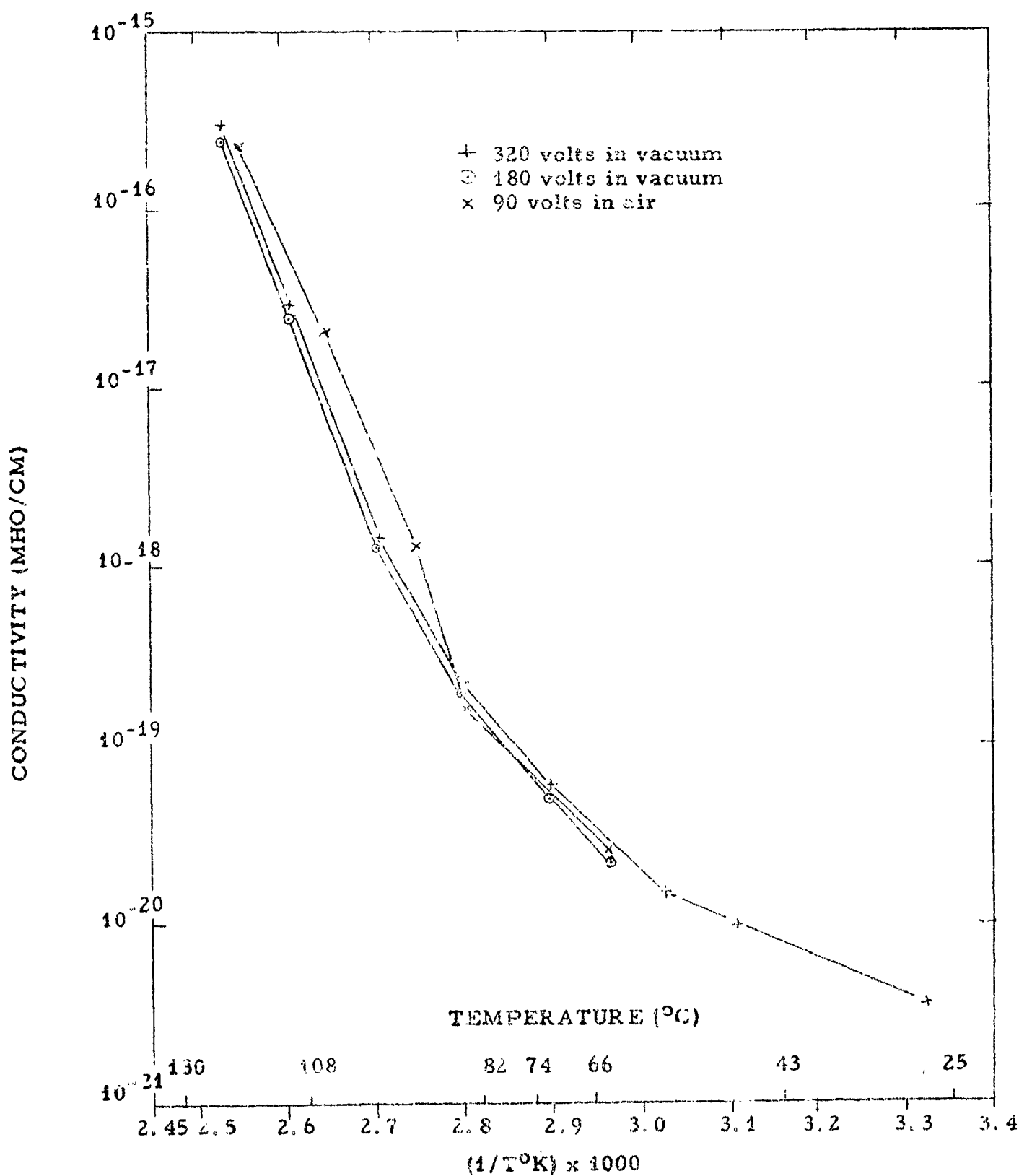


Figure 1. Log $\sigma$  vs 1/T for Mylar Type C after 150 hours conditioning at 125°C

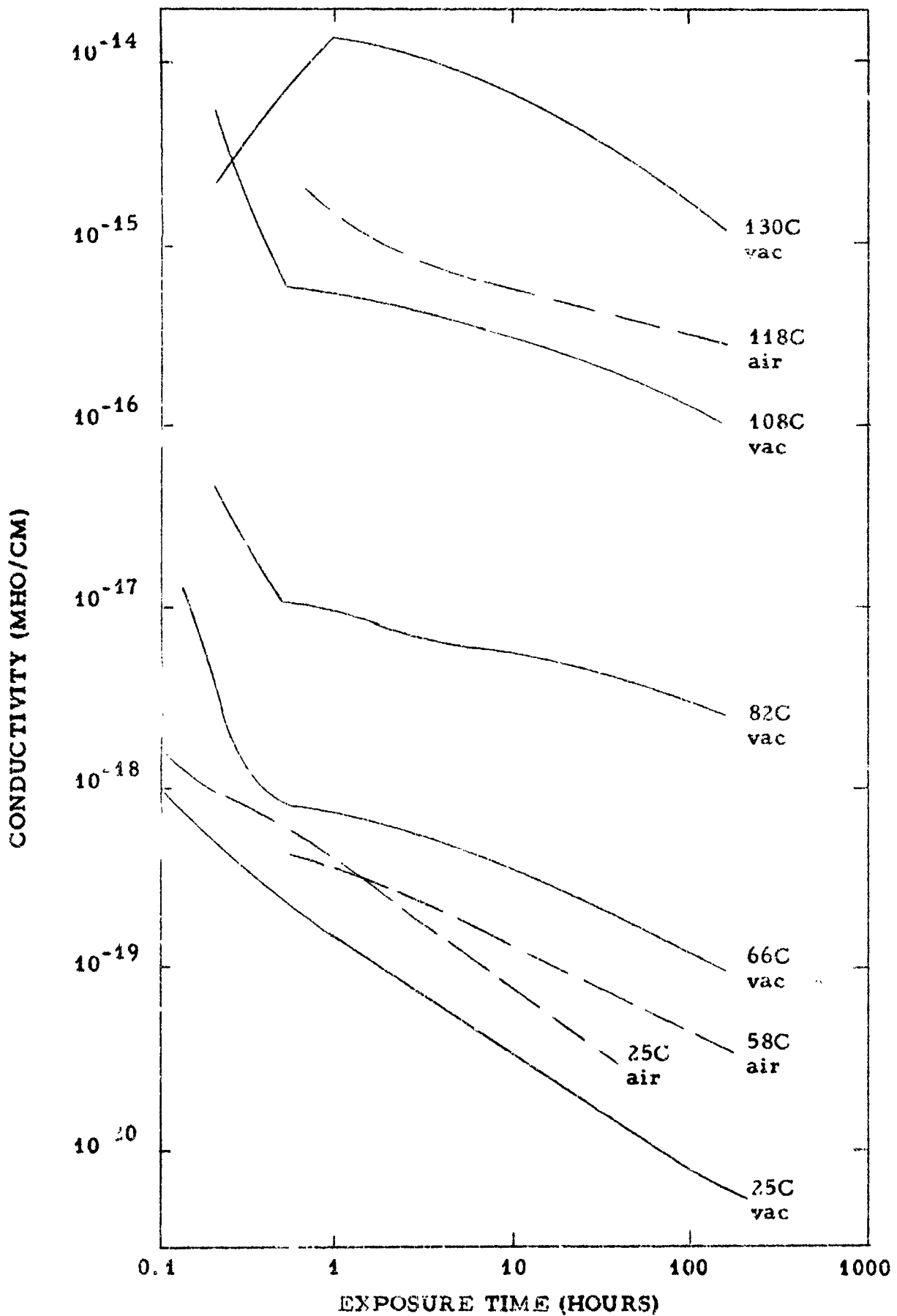


Figure 2. Conductivity of Mylar-C during exposure at temperatures indicate Voltage applied: 320 volts in vacuum; 90 volts in air. Specimens

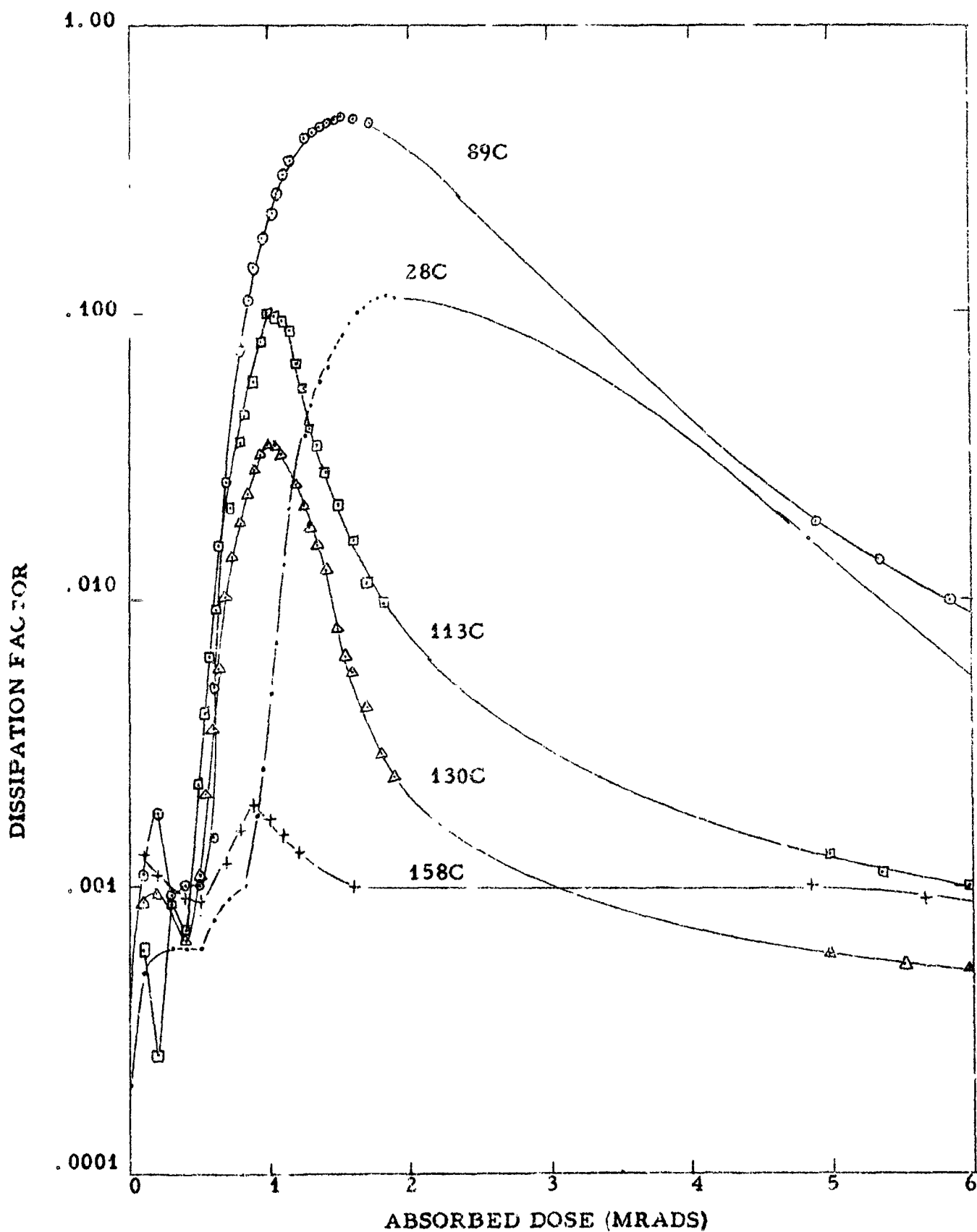


Figure 3. Effect of temperature and absorbed dose on 100 cps  $\tan\delta$  of TFE-7; 30 mil machined specimens. Dose rate 0.2 Mrads per hour.

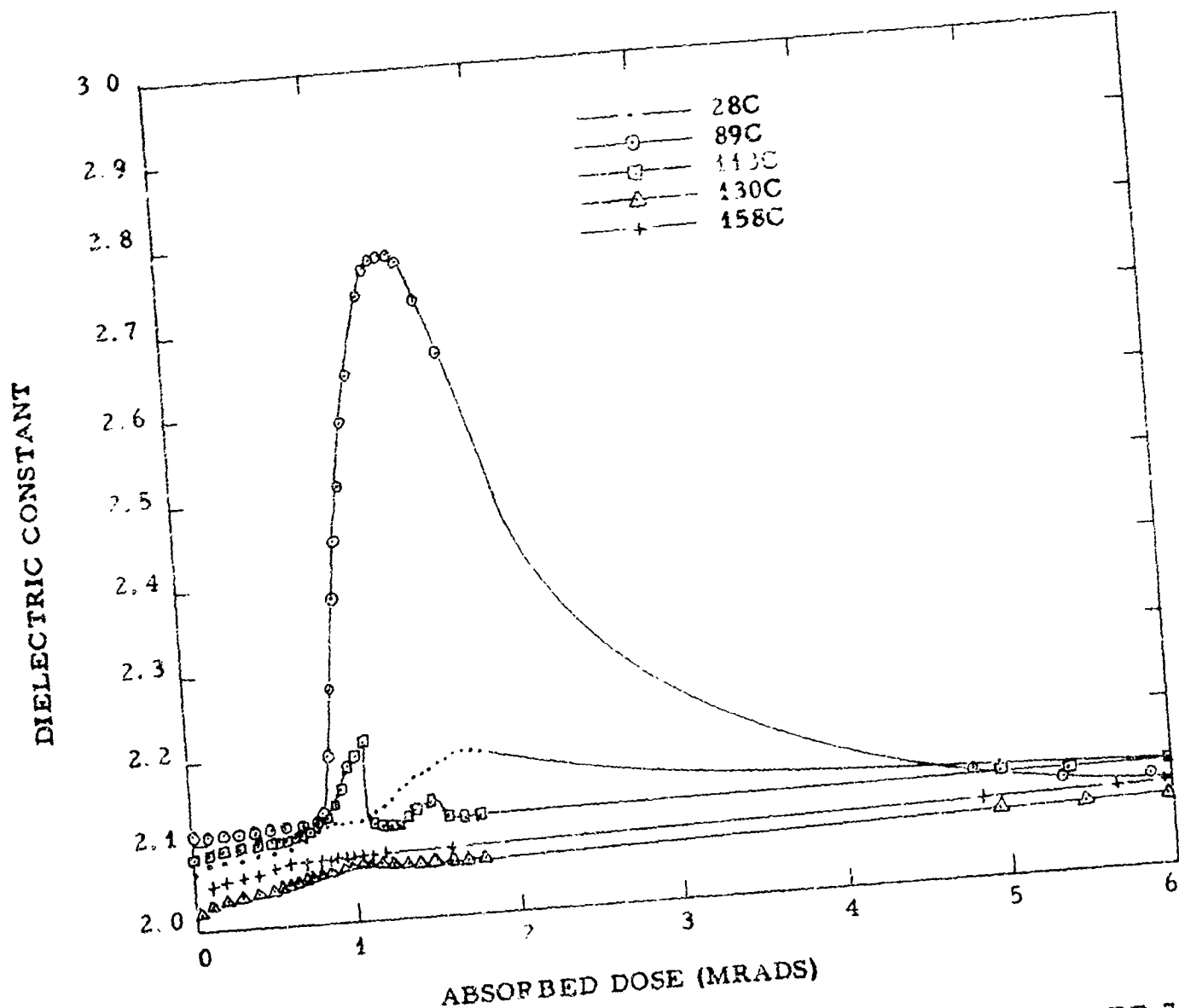


Figure 4. Effect of temperature and absorbed dose on 100 cps  $\epsilon'$  of TFE-7; 30 mil machined specimens. Dose rate 0.2 Mrads per hour.

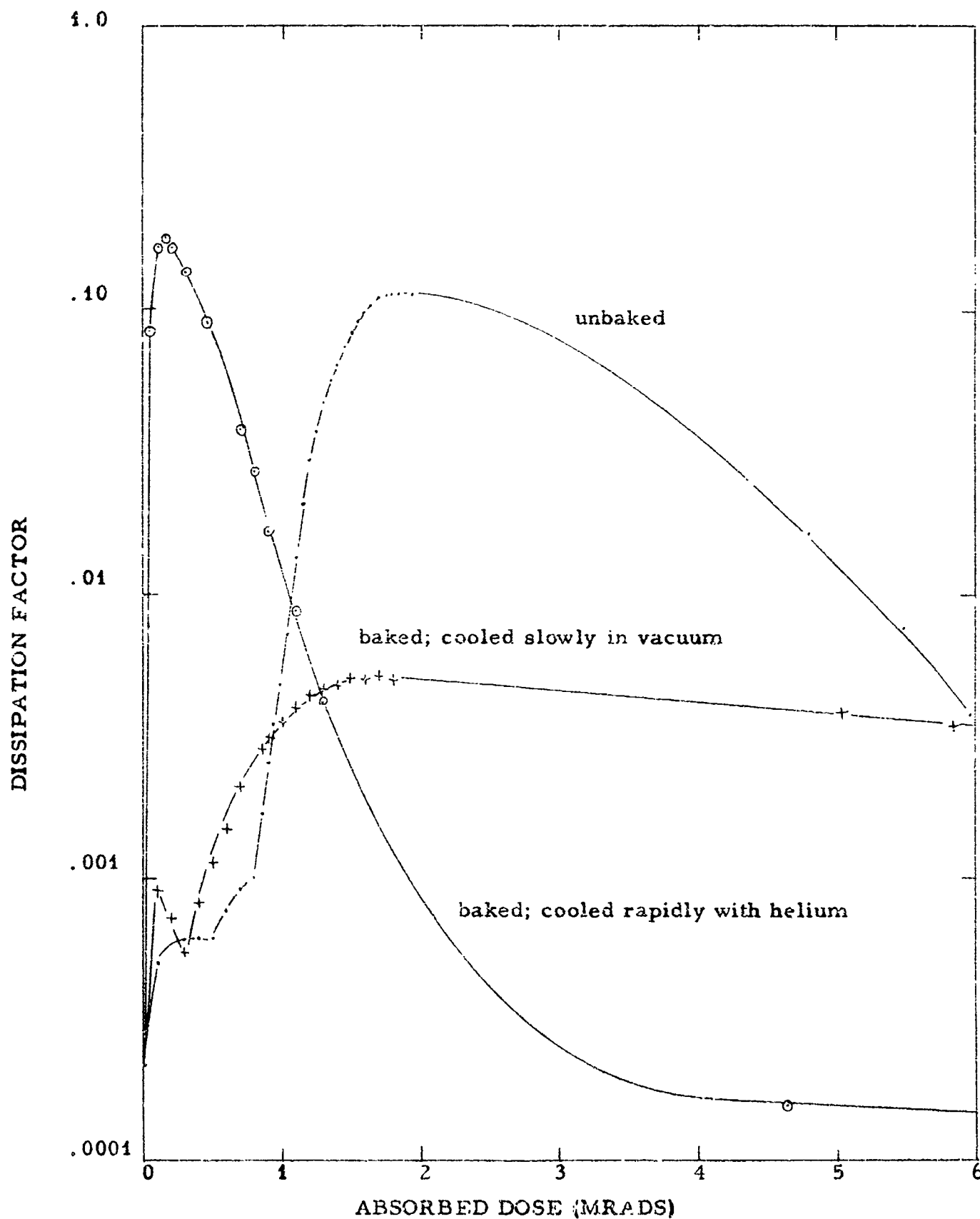


Figure 5. Effect of degassing and absorbed dose on 100 cps  $\tan \delta$  of TFE-7 at room temperature. 30 mil machined specimens. Dose rate 0.2 Mrad per hour.

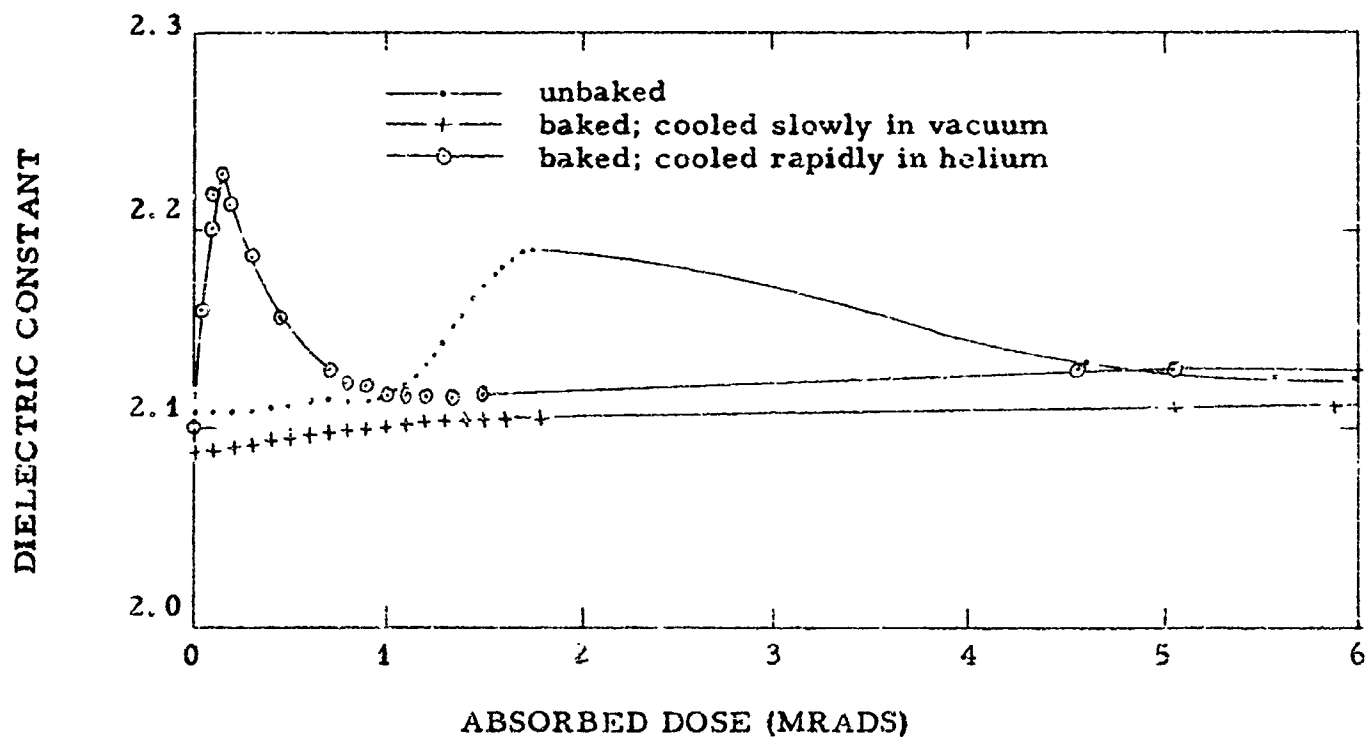


Figure 6. Effect of degassing and absorbed dose on 100 cps  $\epsilon'$  of TFE-7 at room temperature. 30 mil machined specimens. Dose rate 0.2 Mrad per hour.

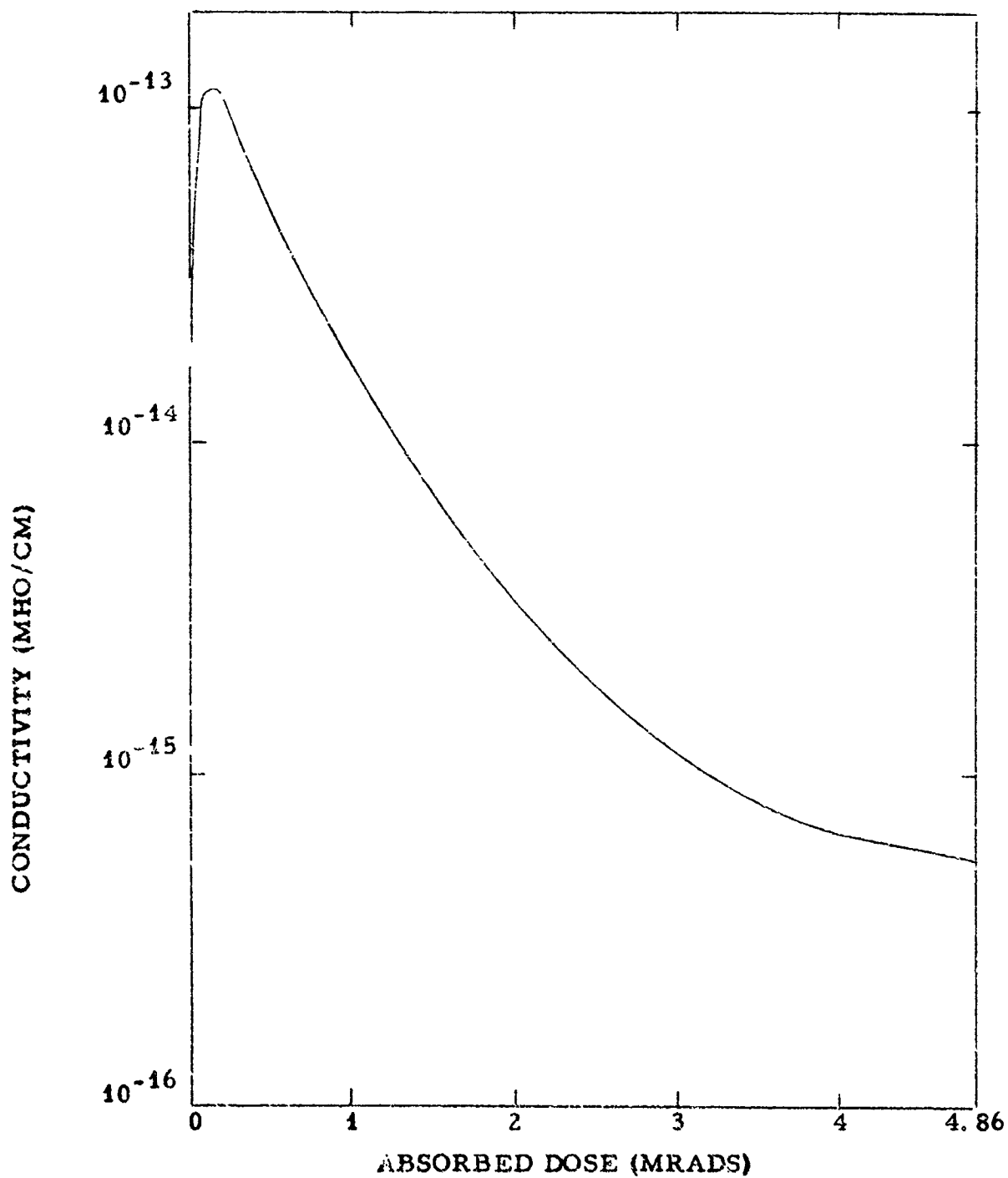


Figure 7. Effect of absorbed dose on induced conductivity of TFE-7 in vacuum at room temperature. Dose rate 0.2 Mrads per hour.



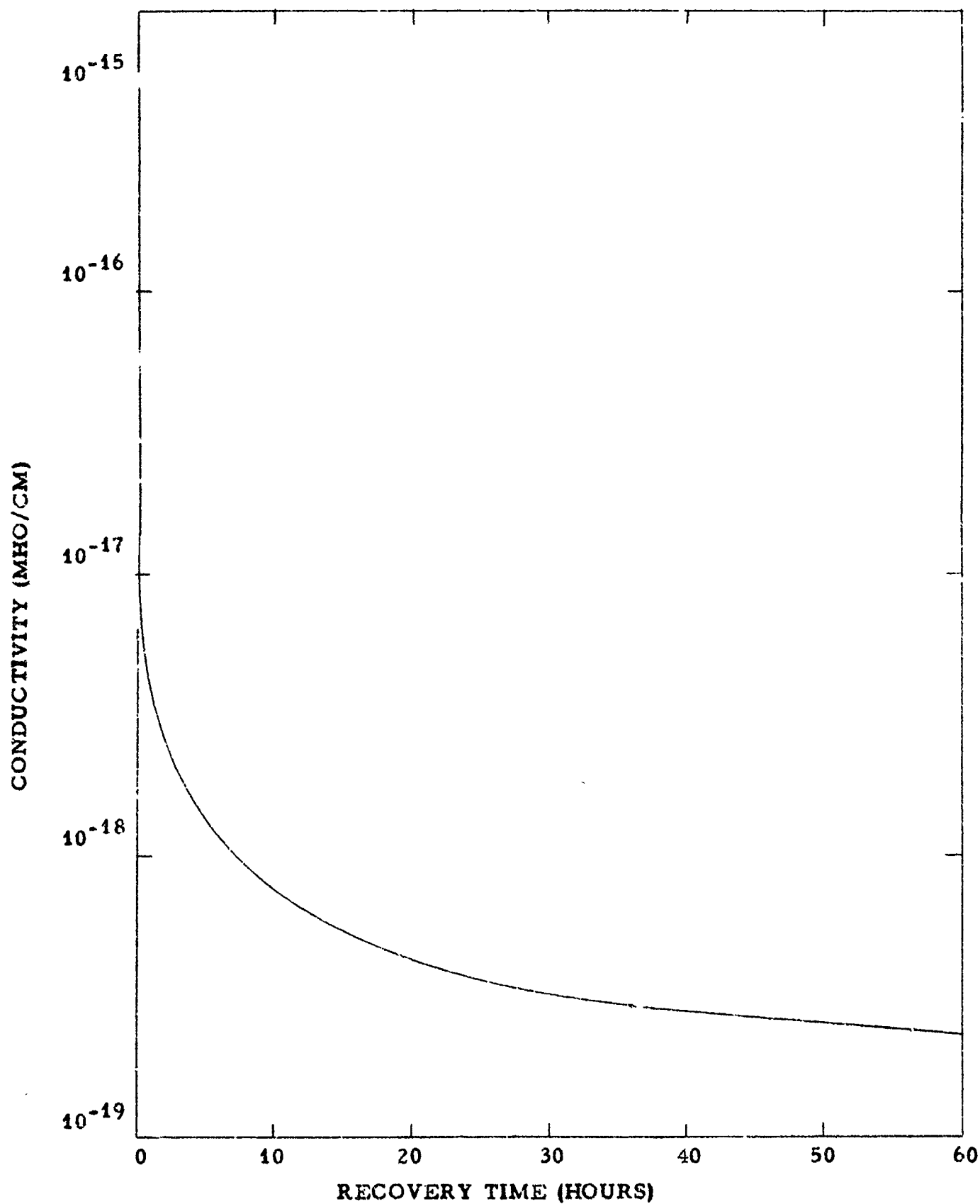


Figure 8. Conductivity of TFE-7 during vacuum recovery following x-ray exposure as shown in Figure 7.